# COAL FINES COMBUSTION IN FLAME REACTION ZONE

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## ABSTRACT

*Upon heating, coal particles devolatilize with subsequent formation of char; oxidation of devolatilized gases, mainly $CH_4$, $H_2$ and other light gases; oxidation of the resulting char. Although these stages overlap in fundamental studies related to coal combustion, it is possible to treat these independently with some modification.*

*Based on the energy balance for combusting coal particles, it is possible to intuitively translate the consequence of coal surface temperature. As a result of exothermic chemical reaction at the particle surface, the ensuing temperature can exceed that of the ambient gas. Due to energy loss by radiation; the effects of heating may be obscured, in which case the particle temperature can be less than that of the gas. The knowledge of the particle temperature is important as it affects many of the stages of reaction occurring during coal combustion and accurate determination of coal surface temperature is necessary as it is a precursor for which practical combustors might be designed and controlled.*

*Devolatilisation and some reactions of the light hydrocarbons are mainly endothermic. During combustion, exothermic chemical reactions predominates both in the surrounding gas and at the particle surface. Due to super equilibrium of reactive transient gases, O, OH and H the particle heating in this zone is greatest. The present paper addresses these observations on both experimental and theoretical basis. These form the pillars for predicting carbon particle temperatures under combustion condition.*

## INTRODUCTION

Temperature attained by combusting coal particles is important for a number of reasons. The particle temperature affects the surface reactions, including the rate of devolatilisation, and its accurate measurement is necessary for an understanding of coal, soot and char combustion. It is also a possible means by which practical combustors might be controlled.

Although devolatilisation and some reactions with carbon are endothermic, exothermic chemical reactions occur both in the surrounding gas and at the particle surface. Porosity increases the available surface for reaction and, consequently, the particle temperature. Graphite particles injected into lean methane-air flat laminar flame have shown that the heating of the particles by chemical reaction at the surface is greatest in the reaction zone of the gaseous flame. This is due to super-equilibrium concentrations of highly reactive species O, H and OH in that zone [1].

These findings are deployed to predict theoretically the carbon particle temperatures to be expected under a variety of conditions.

## EXPERIMENTAL SET-UP

The detailed experimental set up is explained elsewhere [1] and the apparatus is shown schematically in Fig.1. Graphite particles with a mean diameter of 4 µm were introduced into laminar, premixed methane-air flat flames of equivalence ratio, $\emptyset = 0.70$, supported on a flat flame burner at 0.160 atmos. pressure. Gas flow through an elutriator of graphite particles entrained some of them into the premixed methane-air flow. The velocity profiles of particles through the flame, were measured by laser Doppler anemometry. The same equipment gave the particle size distribution from measurements of the modulation, or visibility, of the scattered Doppler signal. The particle number density was obtained from particle count rates with the same laser unit. Gas temperatures through the flame were measured by silica coated fine wire thermocouples of platinum - 20% rhodium v platinum - 40% rhodium, with a correction for radiation loss. Particle temperatures were obtained by the two-colour spectroscopic emission technique, at wavelengths of 0.426 µm and 0.563

μm.

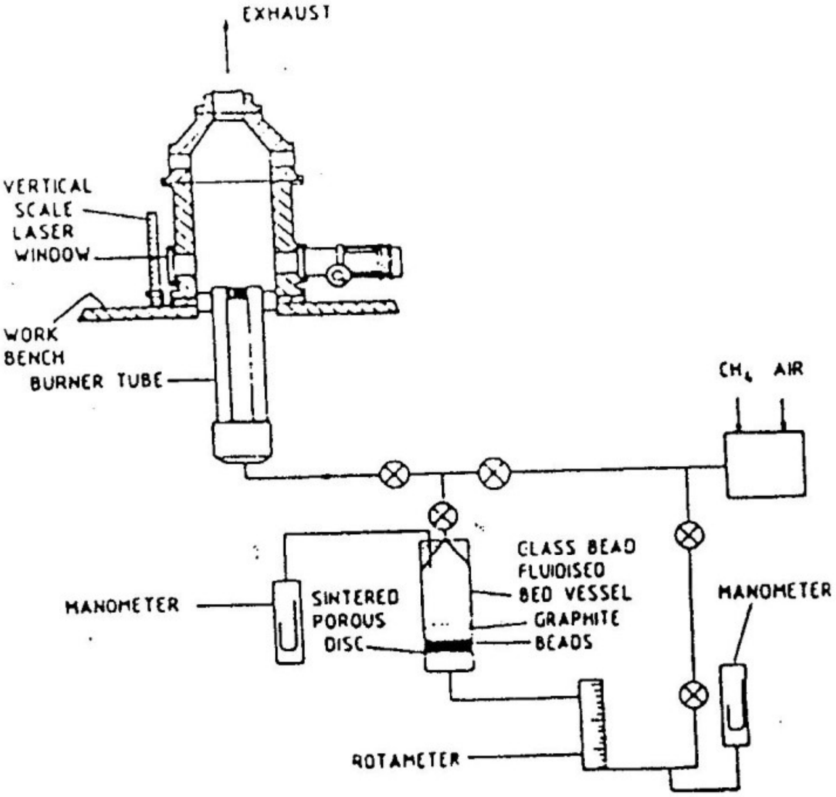

Fig. 1: Apparatus

## RESULTS

Results for a ratio of graphite to gaseous mass of 1.087 x $10^{-3}$ is shown in Fig. 2. If all the graphite were to have been oxidised by $O_2$ to $CO_2$ it would have used only 3.7% of the excess $O_2$ in the burnt gases. Thus the presence of the graphite, is unlikely to create a major perturbation of the methane oxidation kinetics that were employed to explain the observed phenomena. The graphite conservation equation,

$$\dot{\rho}_c = -\frac{d}{dx}(U\rho_c) \tag{1}$$

in conjunction with the measurements, yielded the volumetric mass rate of graphite burning, $\dot{\rho}_c$ along the length of the flame. The gas velocity is u and $\rho_c$ is the graphite mass density per unit volume of mixture. In Fig. 2, the particle and gas temperatures are $T_p$ and $T_g$, respectively.

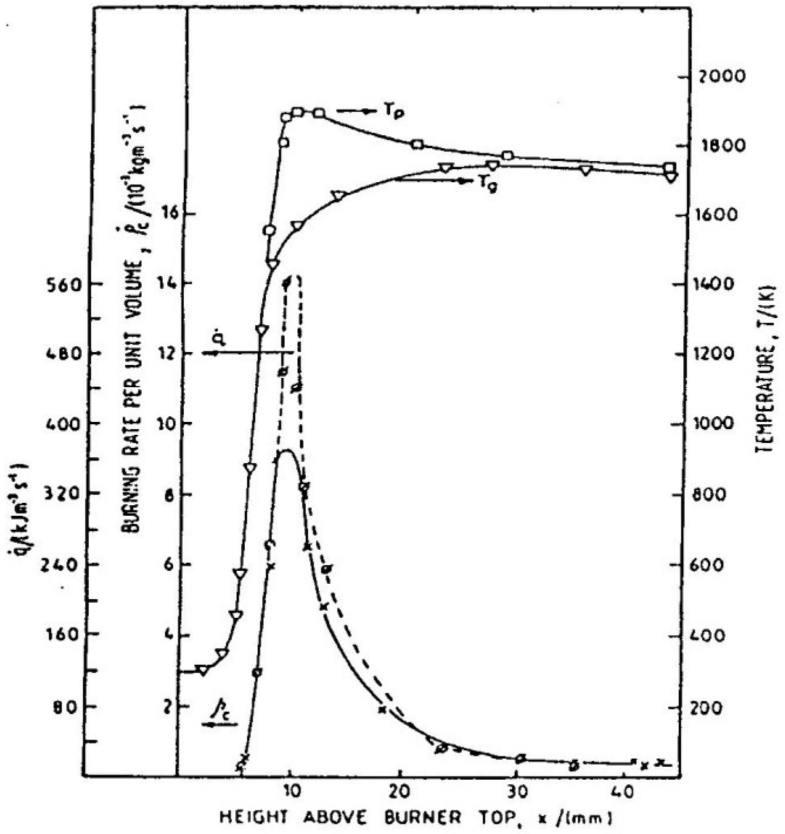

**Fig 2** Graphite mass burning rate and heat release rate through a flat laminar $CH_4$ - air flame. Equivalence ratio, Ø=0.70, pressure = 0.160 atm. Gas and particle temperature also shown

The chemical heat release rate per unit spherical external area of particle, q, can be obtained from the particle energy conservation equation

$$q = h(T_p - T_g) + \sigma(\varepsilon T_p^4 - \alpha T_w^4) + \frac{1}{6}\frac{\delta(\rho d C T_p)}{\delta t} \qquad (2)$$

where; the convective heat transfer coefficient, $h$, was obtained on the assumption of a Nusselt number of 2, $\sigma$ is the Stefan-Boltzmann constant, $\varepsilon$ is the particle emissivity, assumed to be unity, $\alpha$ is its absorptivity to radiation from black surrounds at temperature $T_w$, whilst $\rho$, $d$ and $C$ are the particle density, diameter and specific heat, and $t$ is time. Values of $q$ obtained from experimental conditions, were multiplied by the total external area in unit volume of mixture to yield the volumetric heat release rate q from the combustion of graphite as shown in Fig. 2.

From Fig. 2, the highest rates of graphite burning occur where the gaseous flame reaction rates are highest. This suggests that the highly reactive O, H and OH radicals make an important contribution.

## Gas Phase and Carbon Kinetics

The detailed structure of the gaseous flame was computed from the reaction kinetics and transport data of Dixon-Lewis and Islam [2] and Dixon-Lewis [3]. These yielded profiles of the concentrations of the transient species, O, H and OH, as well as the more stable species $O_2$, $CO_2$, $H_2O$ and $H_2$. Profiles of mole fractions of O, OH and $O_2$, are shown in Fig. 3. The reaction rate coefficients in all cases are of the form

$$k_i = A_i T^{B_i} \exp(-\frac{E_i}{RT}) \qquad (3)$$

where $k_i$ is the specific reaction rate [kg $m^{-2}$ $s^{-1}$], $A_i$ the pre- exponential factor, $T$ the absolute temperature, $E_i$ the activation energy and $R$ the universal gas constant. Values used in the course of the present work are those used by John [4].

$k_i$ in equation (3) is referred to all the available area, including the contribution from internal pores. This creates problems as the internal porosity is, in general, not precisely known. A factor f to express the ratio of the total reactive area to the external surface area shall be introduced,

$$f = \eta \gamma \rho A_g \tag{4}$$

Here $\eta$ is an effectiveness factor equal to the actual reaction rate per particle divided by the rate in the absence of pore resistance to the species, $\gamma$ is the average ratio of particle volume to external surface area (obtained from the size distribution) and $A_g$ is the specific surface area in $m^2g^{-1}$.

The intrinsic burning rate of the graphite, based on the total surface area, from each species at a temperature that was the mean of the gas and particle temperatures was evaluated. The total contributions from $CO_2$, $H_2O$, $O_2$ and $H_2$ are shown by the dashed curve in Fig. 3 and the total contribution from O, H and OH by the dotted curve.

Theoretical heat release rates at the graphite surface were obtained by multiplying the mass burning rate for the appropriate step by the associated heat of reaction. Enthalpy changes $\Delta H$ ($kJg^{-1}$ of C burnt) for a number of possible surface reactions from Habik [5] are given in Table 1. These were used, together with an average enthalpy change $\Delta H = -30$ $kJg^{-1}$ of carbon for attack by OH.

## Devolatilisation and Oxidation of Char

In practical situations, conditions are somewhat different from the controlled laboratory experiments just presented. Nevertheless, there is some basis for a model that assumes that char is oxidised in flame gases originating from earlier devolatilisation and that the carbon surface reactions do not greatly perturb the gas phase kinetics. For coal particle sizes of about 80 μm diameter and higher, Seeker *et al.* [6] have observed that the release of volatiles occurs in jets that have long diffusion and mixing times. Thus pyrolysis reactions occur over longer times.

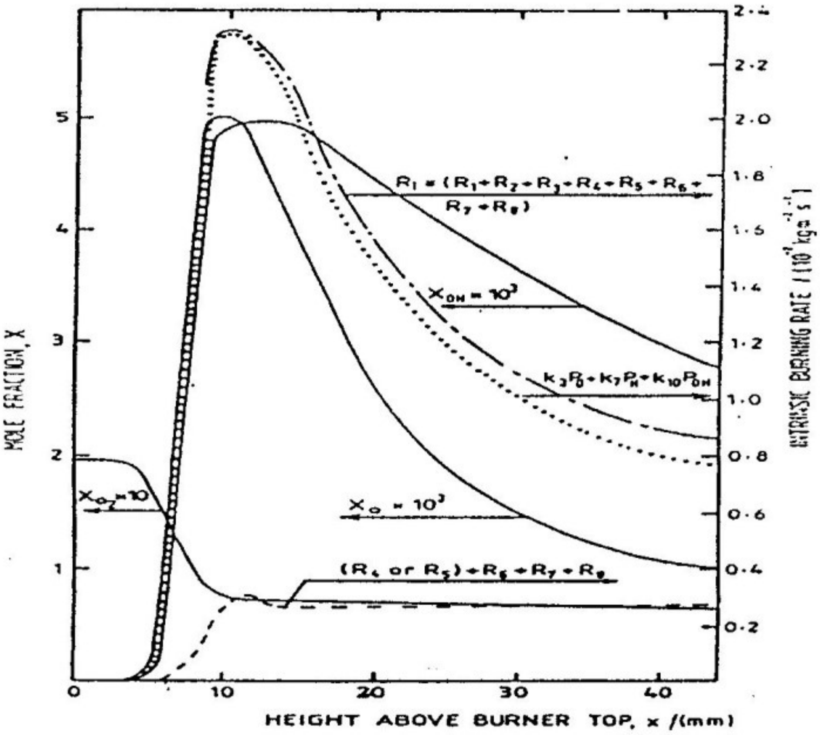

Fig. 3 Computed mole fractions X of O, OH, and $O_2$ for flame of Fig. 2 and burning rates of different species

Table 1 Elementary reactions involved in flame oxidation of carbon particles

| | Reaction | ΔH |
|---|---|---|
| (1) | $C + O \longrightarrow CO$ | -28.7 |
| (2) | $C + OH \longrightarrow CO + H$ | + 6.8 |
| (3) | $C + 0.5O_2 \longrightarrow CO$ | - 8.0 |
| (4) | $C + H_2O \longrightarrow CO + H_2$ | +12.1 |
| (5) | $C + CO_2 \longrightarrow 2CO$ | +16.4 |
| (6) | $C + H_2 \longrightarrow CH_2$ | +32.1 |

However, for smaller sizes there seemed to be a more effective early mixing and oxidation of volatiles. Hertzberg *et al.* [7] found that for sizes less than about 50 µm, devolatilisation was so rapid that the overall combustion process was controlled by chemical reaction and diffusion in the gas phase.

Bradley and Lee [8] have suggested that flame propagation rates in dusts are controlled largely by gas phase kinetics. Particle sizes of less than 1 µm diameter can be entirely oxidized in the gaseous reaction zone by active species [5, 8]. With larger particles there will be some reaction in this zone, followed by continued, slower, reaction in hot gases much closer to equilibrium conditions. For the very small sizes it is as if the solid carbon were burning as a gas. As particle size increases, reaction becomes increasingly controlled by the diffusion rate of the reacting species to the carbon surface and less by the chemical reaction rate.

## Computation of Particle Temperatures

The temperatures attained by carbon particles in a $CH_4$ - air mixture of equivalence ratio of 0.84 for a temperature of the reactants of 298 K and a pressure of 1 atmos will be considered. The mixture is an approximate simulation of the combustion of coals of high volatile content, [4].

Fig. 4 shows computed values of u and $T_g$ and of q and $T_p$ for initial particle diameters of 0.4, 4, 40 and 400 µm. The initial value of f was 1.0 and $\eta$, $\gamma$ and $\rho$ were assumed constant during the particle burning. It was further assumed that no radiation was received by particles from the cooler surrounding wall.

Initially, with constancy in the values of f, $\eta$ , $\gamma$ and $\rho$ it is observed that the change in the heating rate is greatest in the reaction zone. Also the highest heating rate is observed at the highest particle temperatures. To be inferred is that particles in the size range more than 400 µm has the least heating rate effect. Further, the gas temperature of the mixture in question is nearly the same with the particle temperature for carbon particles with 0.4 µm diameter. In addition, the general observation on the gas velocity profile is obtained in which it is the same as the gaseous temperature profile.

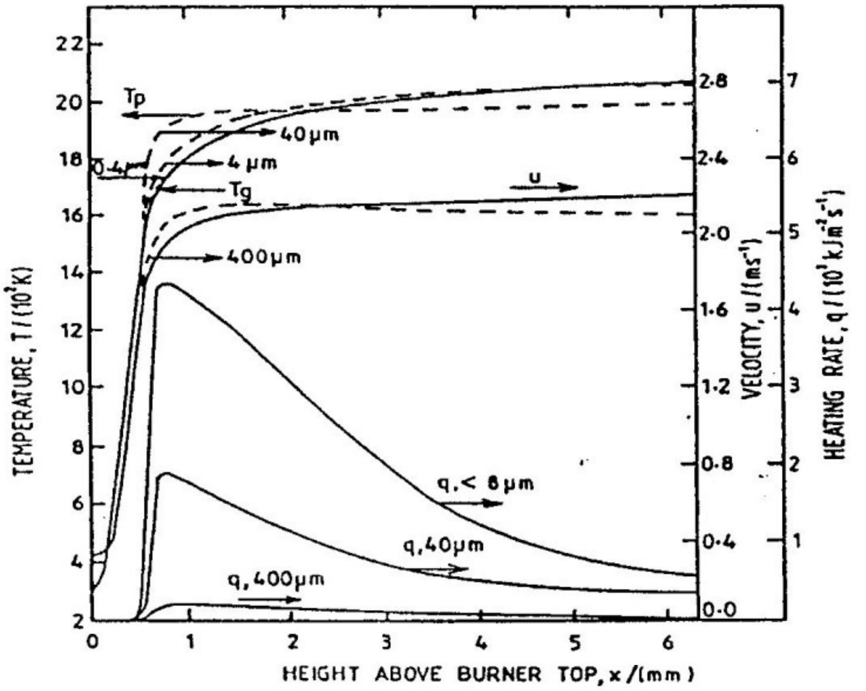

**Fig. 4 Computed values of u, $T_g$, $T_p$ and particle heat release rate, q, per unit external surface area for three different particle diameters. Equivalence ratio, = 0.84, pressure = 1.0 atmos., f = 1, $T_w$ = 3000 K.**

## Particle Heating in the Flame Reaction Zone: No Radiation from Surroundings

The effect of f shall be considered, at the position in the flame where the concentration of OH is a maximum. The gas temperature here is 1720 K, with a 4.5% mole fraction of $O_2$. Figure 5 shows value of q as a function of particle diameter when f = 1, 10 and 100. In the chemically limited regime, an increase in porosity creates more surface area for reaction, with the result that q increases.

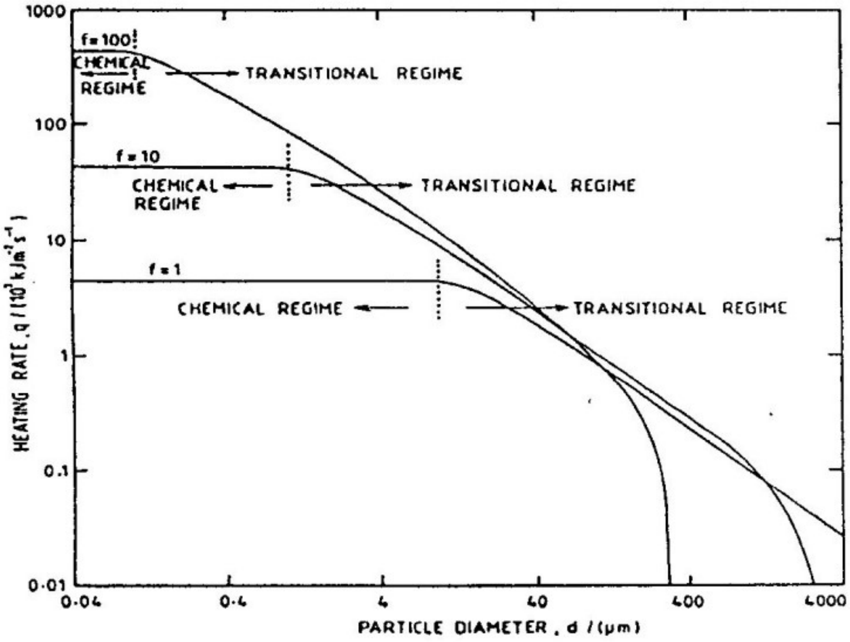

**Fig. 5 Variation of q with particle diameter at maximum OH concentration for three different values of f (Logarithmic scale).**

At a given value of f, as d increases eventually diffusion becomes rate limiting, and q begins to fall. Ultimately, the heating rate is diffusion-limited and inversely proportional to $d^2$. On the other hand, an increase in d also gives rise to a decrease in the convective heat transfer coefficient, h. This reduces the rate of heat loss by convection, $H_c$, the first term on the right hand of equation (2), and the particle temperature initially increases, as shown in Fig. 6. With further increase in d, eventually diffusion limitations become appreciable and the associated decrease in q results in a decrease in particle temperature. The fall in $H_c$ with increase in d, due to the decrease in h, is stronger even than the decrease in q due to diffusion

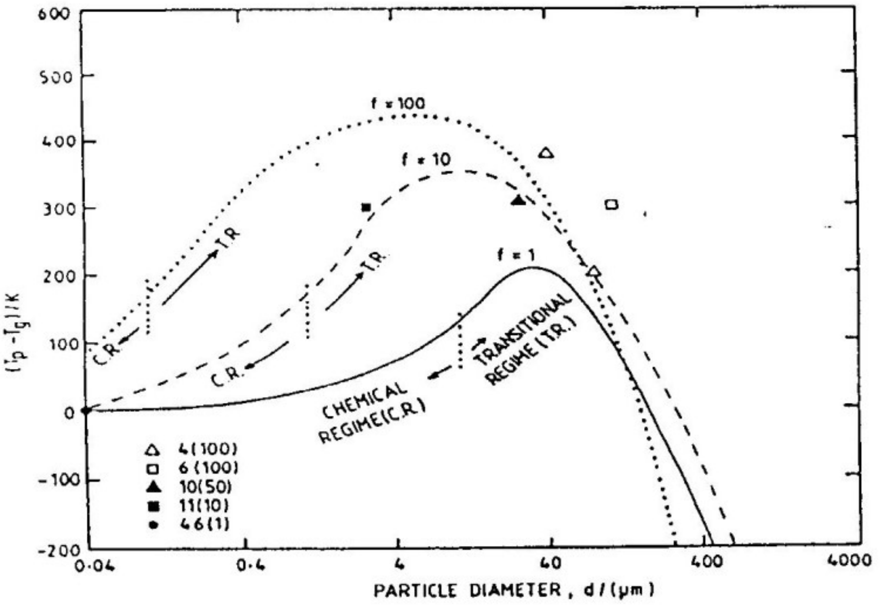

**Fig. 6 Particle temperature increase above gas temperature at different particle diameters for three different values of f. Position of maximum OH concentration. Experimental points shown: figures in brackets on key show approximate value of f.**

The decrease in the ratio $H_c/q$ for the three different values of f is shown in Fig. 7. When the ratio becomes zero, any further increase in diameter results in $T_p$ becoming less than $T_g$. With the accumulation term in equation (2) neglected, the equation becomes

$$1 = \frac{H_c}{q} + \frac{H_r}{q} \tag{5}$$

Clearly, a decrease in $H_c/q$ is associated with an increase in the radiative term, $H_r/q$. In Fig. 6, the values of f were taken to be close to unity for soot and in the region of 100 for coals. Reaction zone experimental data are also plotted in Fig. 6. Although a variety of experimental conditions are represented, different from those of the present theory, there is, nevertheless, some general support for the theoretical predictions.

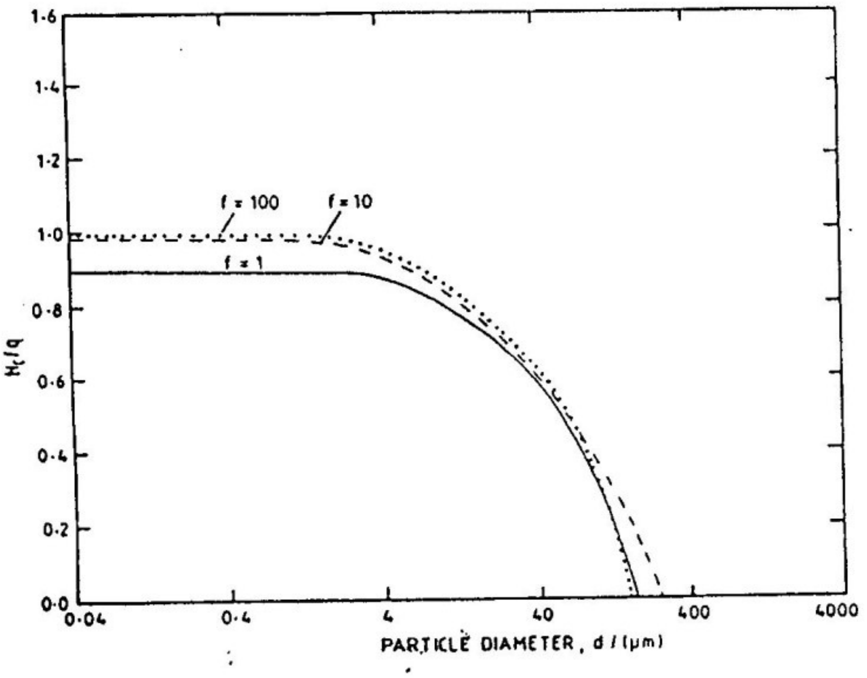

**Fig. 7 Variation of $H_c/q$ with particle diameter for three different values of f. Position of maximum OH concentration.**

## Particle Heating in the Flame Reaction Zone: $T_g = T_w$

Because of the radiative energy exchange between the walls and the particles, the wall temperature is an important variable. The calculations of Section 5.1 were repeated with the sole change that $T_w$ was equal to 1720 K and $\alpha$ was assigned a value of unity. There was little change in the value of q for the different particle diameters. The corresponding variations of $(T_p - T_g)$ and $H_c/q$ with particle diameter are shown in Figs. 8 and 9 respectively.

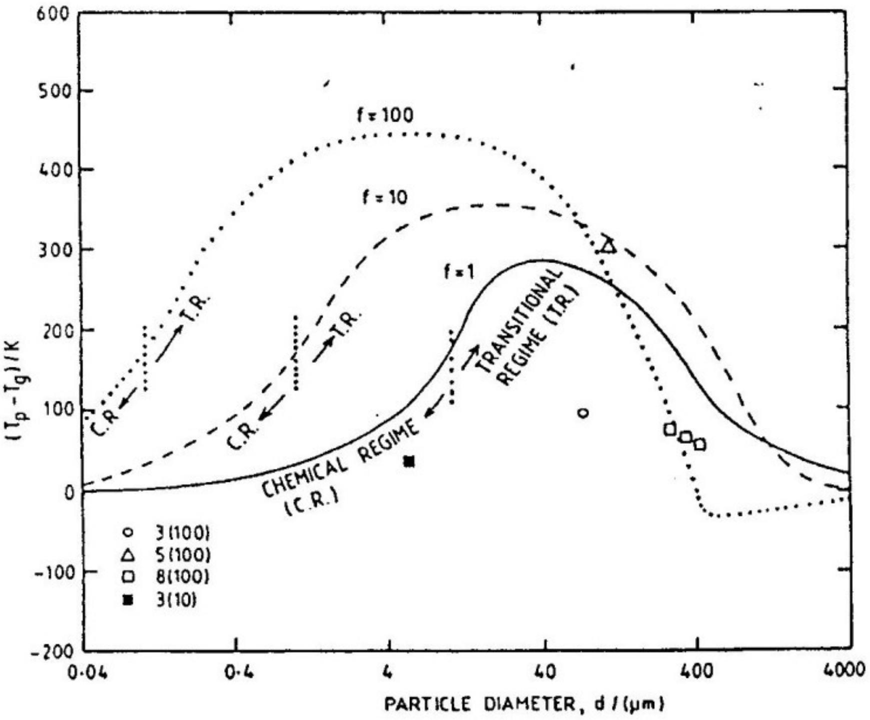

Fig. 8 **Particle temperature increase above gas temperature at different particle diameters for three different values of f. Position of maximum OH concentration, $T_g = T_w$. Experimental points shown: figures in brackets on key show approximate value of f.**

The values of $(T_p - T_g)$ are increased when there is a radiative contribution from the walls. Fig. 9 shows higher values of $H_c/q$ than does Fig. 7 for the same diameter. Fig. 8 shows $T_p > T_g$ in the region of 400 μm diameter with $T_g = T_w$ and f = 1 and 10, whereas with no radiation from the surroundings Fig. 6 shows $T_p < T_g$.

Experimental data are scarce than for the previous conditions, Fig. 8. It would be expected for these conditions that as q decreases with increase in diameter, so the value of $T_p$ would tend to that of $T_g$, and this is confirmed by Fig. 8.

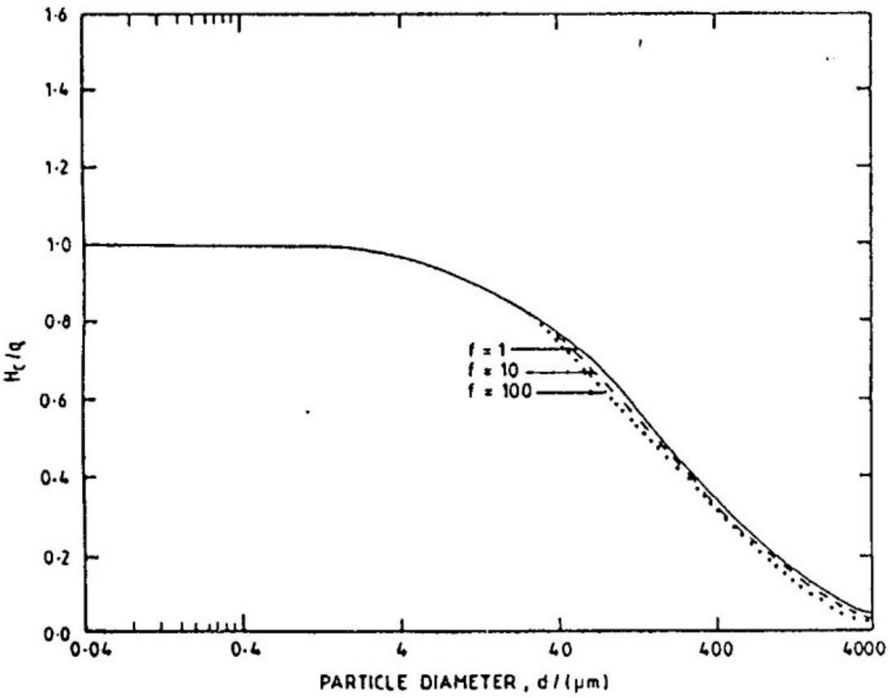

**Fig. 9 Variation of $H_c/q$ with particle diameter for three different values of f. Position of maximum OH concentration, $T_g = T_w$.**

## CONCLUSIONS

(i) Experimental studies of the heating of graphite particles in a low pressure $CH_4$ - air laminar flame have been reported. The heating is explained by the reactions of the gas phase species on the surface of the solid.

(ii) From the computed structure of atmospheric $CH_4$ - air flames it is possible to estimate the temperature of char particles in more practical coal flames. Where comparable experimental data are available, comparisons are made with these. Chemical and diffusion controlled regimes have been identified for the different species.

(iii) The greatest elevations of particle temperature above gas temperature occur in the gaseous flame reaction zone and are attributable to the exothermic reactions of O, H and OH with carbon. The elevations, which can be more than 400 K, increase with particle porosity and are a maximum in the particle size range 1-40 μm.

(iv) As the particle size increases, the energy loss by convection decreases and the particle temperature increases. Eventually, when the particle size has become large enough to give diffusion rate control of species, there is a decrease in the heat release rate at the solid surface and consequently in $T_p$.

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